Charge order, metallic behavior and superconductivity in $La_{2-x}Ba_xCuO_4$ with $x = 1/8$

C. C. Homes,* S. V. Dordevic,† G. D. Gu, Q. Li, T. Valla, and J. M. Tranquada
Condensed Matter Physics & Materials Science Department,
Brookhaven National Laboratory, Upton, New York 11973

(Dated: February 6, 2008)

The *ab*-plane optical properties of a cleaved single crystal of $La_{2-x}Ba_xCuO_4$ for $x = 1/8$ ($T_c \simeq 2.4$ K) have been measured over a wide frequency and temperature range. The low-frequency conductivity is Drude-like and shows a metallic response with decreasing temperature. However, below $\simeq 60$ K, corresponding to the onset of charge-stripe order, there is a rapid loss of spectral weight below about 40 meV. The gapping of single-particle excitations looks surprisingly similar to that observed in superconducting $La_{2-x}Sr_xCuO_4$, including the presence of a residual Drude peak with reduced weight; the main difference is that the lost spectral weight moves to high, rather than zero, frequency, reflecting the absence of a bulk superconducting condensate.

PACS numbers: 74.25.Gz, 74.72.-h, 78.30.-j

It has been proposed that charge inhomogeneity, especially in the form of stripes, is a phenomenon intrinsic to doped antiferromagnets such as cuprate superconductors [1, 2, 3, 4, 5]. Static spin and charge stripe order has been observed by diffraction in a couple of cuprate compounds [6, 7, 8]; however, it appears to compete with superconductivity [7]. The electronic structure of the stripe-ordered state remains to be clarified experimentally. This is an important issue, as it bears on the question of whether dynamic stripes might be compatible with superconductivity, and possibly even necessary for the high transition temperatures [9]. Kivelson, Fradkin, and Emery [10] argued that straight, ordered stripes should develop a charge-density-wave (CDW) order along the stripes, which would compete with pairing correlations. Castellani *et al.* [11] have suggested that such commensurate stripes should be insulating. Previous investigations of stripe correlations using infrared reflectivity on oriented, cut-and-polished crystals of $La_{1.88-y}Nd_ySr_{0.12}CuO_4$ (LNSCO) [12, 13] and $La_{2-x}Sr_xCuO_4$ (LSCO) [12, 14] have resulted in a certain amount of controversy [15, 16].

In this Letter we report on the *ab*-plane optical properties of a *cleaved* single crystal of $La_{2-x}Ba_xCuO_4$ (LBCO) for $x = 1/8$. The same crystal was used in a recent soft-X-ray resonant-diffraction study that demonstrated charge order below 60 K [8]. Working with a cleaved surface greatly reduces the possibility of inadvertent surface misorientation that can result in spurious effects [16]. The optical conductivity initially shows a normal (for the cuprates) metallic response with decreasing temperature. Below $\simeq 60$ K, the conductivity at very low frequency appears to either remain roughly constant or increase slightly as the temperature is reduced; however, there is a general loss of spectral weight (the area under the conductivity curve) below $\simeq 300$ cm^{-1}, a consequence of the substantial reduction in the density of carriers. The missing spectral weight is redistributed to higher frequencies, and is fully recovered only above about 2000 cm^{-1}. Coinciding with the depression of the carrier density, the transition to superconductivity is depressed to 2.4 K. The gapping of the single-particle excitations looks remarkably similar to that recently reported for superconducting $La_{1.85}Sr_{0.15}CuO_4$ [17], which is known to have a *d*-wave gap [18]. The presence of a residual Drude peak indicates that charge order is compatible with a "nodal metal" state [19].

Single crystals of $La_{2-x}Ba_xCuO_4$ with $x = 1/8$ were grown by the floating zone method. The sample used in this study had a strongly suppressed bulk $T_c \simeq 2.4$ K as determined by magnetic susceptibility. The sample was cleaved in air, yielding a mirror-like *ab*-plane face. The *ab*-plane reflectance has been measured at a near-normal angle of incidence over a wide temperature and spectral range using an *in-situ* evaporation technique [20]. The optical properties are calculated from a Kramers-Kronig analysis of the reflectance, where extrapolations are supplied for $\omega \to 0, \infty$. At low frequency, a metallic Hagen-Rubens response is assumed ($R \propto 1 - \omega^{1/2}$). Above the highest-measured frequency in this experiment the reflectance of $La_{1.85}Sr_{0.15}CuO_4$ has been employed to about 40 eV [21]; above that frequency a free-electron approximation has been assumed ($R \propto 1/\omega^4$).

The reflectance of $La_{1.875}Ba_{0.125}CuO_4$ is shown from ≈ 20 to 25,000 cm^{-1} in Fig. 1 for a variety of temperatures. The reflectance is typical of many cuprates, with a plasma edge at ≈ 1 eV and a low-frequency reflectance that increases with decreasing temperature, indicative of a metallic response. However, below ≈ 60 K the low-frequency reflectance in the $200-2000$ cm^{-1} region stops increasing and begins to decrease, as shown in more detail in the inset of Fig. 1. Note that below ≈ 180 cm^{-1} the reflectance continues to increase below 60 K. This suppression of the far-infrared reflectance is unusual and has not been observed in other studies of LSCO or LNSCO [12, 14, 17, 21, 22, 23, 24] for $T > T_c$.

The optical conductivity is shown in Fig. 2, with the conductivity between 295 and 60 K in panel (a)

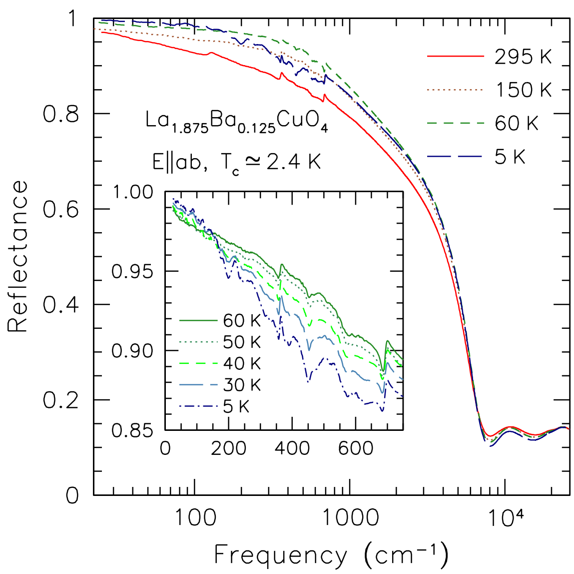

FIG. 1: (Color online) The reflectance at a near-normal angle of incidence for several temperatures over a wide spectral range for a cleaved surface of $La_{1.875}Ba_{0.125}CuO_4$ ($T_c \simeq$ 2.4 K) for light polarized in the a-b plane. The infrared reflectance increases with decreasing temperature until about 60 K, below which it is suppressed in the $200 - 2000$ cm^{-1} region. Inset: The detailed temperature dependence of the far-infrared reflectance at and below 60 K. (The resolution is 2 cm^{-1}.)

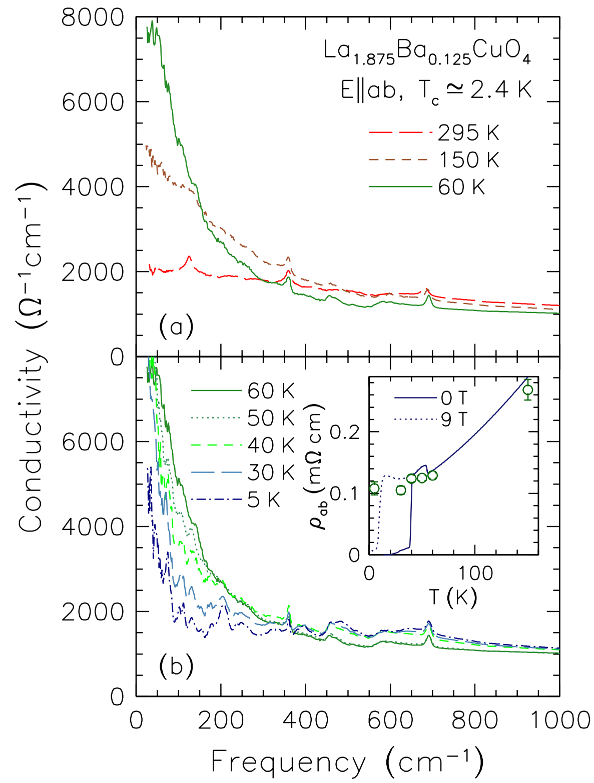

FIG. 2: (Color online) The ab-plane optical conductivity of $La_{1.875}Ba_{0.125}CuO_4$ ($T_c \simeq 2.4$ K). (a) The conductivity in the infrared region between 295 and 60 K, showing a steady narrowing of the Drude-like component. The sharp features in the conductivity are the normally-allowed infrared active vibrations; a feature at $\simeq 125$ cm^{-1} at 295 K is not visible at 60 K. (b) The conductivity for several temperatures below 60 K. A new vibrational feature appears at $\simeq 205$ cm^{-1} below about 40 K. Inset: The ab-plane resistivity of $La_{2-x}Ba_xCuO_4$ for $x \simeq 1/8$ (solid curve) and in an 9 T magnetic field (dotted curve). The open circles are the estimated values for the dc resistivity calculated from the Drude parameters in Table I.

and the behavior below 60 K in (b). The conductivity can be described as a combination of a coherent temperature-dependent Drude component, and an incoherent temperature-independent component. The "two-component" expression for the real part of the optical conductivity is

$$\sigma_1(\omega) = \frac{1}{60}\,\frac{\omega_{p,D}^2\Gamma_D}{\omega^2+\Gamma_D^2} + \sigma_{MIR}, \qquad (1)$$

where $\omega_{p,D}^2 = 4\pi n_D e^2/m^*$ is the square of the Drude plasma frequency, n_D is a carrier concentration associated with coherent transport, m^* is an effective mass, $\Gamma_D = 1/\tau_D$ is the scattering rate, and σ_{MIR} is the mid-infrared component. (When ω_{pD} and $1/\tau_D$ are in cm^{-1}, σ_1 has the units Ω^{-1}cm^{-1}.) The conductivity in the mid-infrared is often described by a series of overdamped Lorentzian oscillators which yield a flat, incoherent response in this region. To simplify the fitting and reduce the number of parameters, a constant background ($\sigma_{MIR} \sim 850\,\Omega^{-1}$cm^{-1}) has been used. The fitted Drude parameter values are summarized in Table I.

Between room temperature and 60 K, the normalized Drude carrier density (last column of Table I) remains constant, while the scattering rate scales with the temperature ($\hbar/\tau \simeq 2k_BT$), similar to what has been seen in the normal state of other metallic cuprates [22, 25]. Below 60 K, a new and surprising behavior occurs: the carrier density decreases rapidly, with a reduction of nearly 75% at 5 K. We note that this behavior is correlated with the onset of charge-stripe order as detected by diffraction techniques [6, 8]. The scattering rate also continues to decrease as n_D drops.

It is interesting to compare with measurements of the in-plane resistivity, ρ_{ab}, shown in the inset of Fig. 2(b), obtained on a sister crystal. On cooling, the decrease in ρ_{ab} is interrupted by a slight jump at the structural transition, at about 54 K [6, 7]. The drop in ρ_{ab} below about 40 K is likely due to filamentary superconductivity [7, 26], as magnetic susceptibility measurements show that the bulk superconducting transition is at 2.4 K. The non-bulk response can be suppressed through the application of a magnetic field (9 T); the resulting curve shows a nearly constant value for the resistivity below 54 K until the onset of bulk superconductivity at low temperature. The Drude expression for the dc resistivity, in units of Ω cm, is $\rho_{dc} = 60/(\omega_{p,D}^2\tau_D)$. The results obtained from the parameter values in Table I, indicated by circles in

TABLE I: Drude parameters from two-component fits to the low-frequency *ab*-plane conductivity of $La_{1.875}Ba_{0.125}CuO_4$. The last column relies on the assumption that m^* is temperature independent.

T (K)	$\omega_{p,D}$ (cm^{-1})	$1/\tau_D$ (cm^{-1})	$n_D(T)/n_D(60\ K)$
295	7010 ± 120	657 ± 35	0.98 ± 0.12
150	7070 ± 80	223 ± 9	0.99 ± 0.06
60	7090 ± 50	108 ± 2	1.00 ± 0.06
50	6560 ± 50	89 ± 2	0.85 ± 0.05
40	5740 ± 40	68 ± 2	0.65 ± 0.04
30	4930 ± 40	43 ± 2	0.48 ± 0.04
5	3690 ± 50	28 ± 2	0.27 ± 0.04

the inset figure, are in good agreement with the transport data. Thus, the flattening out of ρ_{ab} and slight upturn at low temperature corresponds to a decrease in carrier density, and not to an increase in scattering rate.

The optical conductivity lost from the Drude peak must shift to a different frequency range. Where does the missing spectral weight go? To see this, we consider the spectral weight, given by

$$N(\omega_c, T) = \int_{0^+}^{\omega_c} \sigma_1(\omega, T)\, d\omega. \qquad (2)$$

as a function of the cut-off frequency, ω_c. If ω_c is large enough to cover all relevant transitions, then $N(\omega_c)$ should be proportional to the carrier density and independent of the scattering rate; this is known as the f-sum rule. Figure 3 shows $N(\omega)$ for several temperatures above and below 60 K; the inset shows the temperature dependence of $N(\omega_c)$ for three different values of ω_c. As the temperature decreases from 295 K down to 60 K, there is an increase in the low-frequency spectral weight, consistent with results for $La_{2-x}Sr_xCuO_4$ [27]. On cooling below 60 K, the spectral weight below 100 cm^{-1} continues to increase [28]; however, the net spectral weight up to 300 cm^{-1} shows a substantial decrease. The missing spectral weight is finally recovered for $\omega_c \sim 2000$ cm^{-1}, above which $N(\omega)$ is temperature independent. Similar behavior of the spectral weight has been observed in an electron-doped cuprate system [29]. The loss of spectral weight below 60 K suggests a gapping of states near the Fermi level, while the continued presence of a Drude component indicates that some states are not gapped. From these results we infer the presence of an anisotropic charge excitation gap in the stripe-ordered phase.

It has been predicted theoretically that CDW order should compete with superconductivity within an ordered array of stripes [10]. If this were the case, then, as discussed by Zhou *et al.* [30], one would expect the development of a CDW gap within the stripes to occur in the “antinodal” regions of reciprocal space, where the extrapolated Fermi surface has straight, well-nested portions. Note that the antinodal states exhibit a pseudogap in the normal state [18, 31], so that development of a true

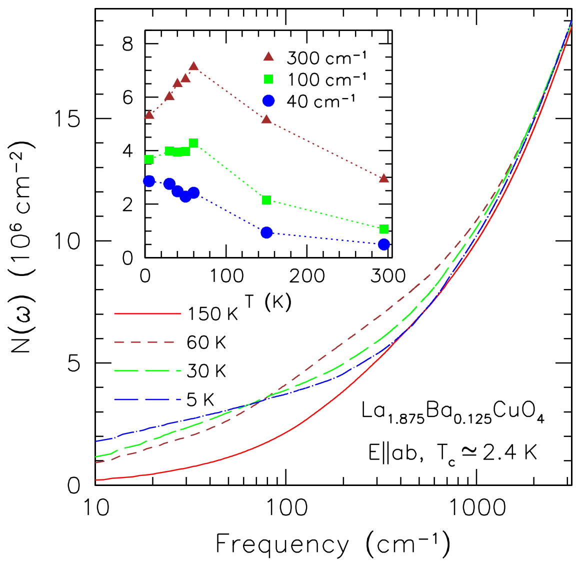

FIG. 3: (Color online) The *ab*-plane spectral weight N(ω) of $La_{1.875}Ba_{0.125}CuO_4$ for several different temperatures above and below 60 K. To simplify the units, the conductivity has been expressed in cm^{-1}. Inset: The temperature dependence of the spectral weight for cut-off frequencies ω_c = 40, 100 and 300 cm^{-1}.

CDW gap would only affect states at finite binding energy.

Alternatively, we find that the decrease in the low-frequency conductivity at low temperature in the stripe-ordered state of $La_{1.875}Ba_{0.125}CuO_4$ looks surprisingly similar to that found in the superconducting state of $La_{1.85}Sr_{0.15}CuO_4$ [17], both in terms of energy scale and magnitude. A residual Drude peak was also observed in the latter superconductor. This similarity suggests an intimate connection between the charge gap of the stripe-ordered state and the pairing gap of the superconductor. Could the charge gap correspond to pairing without phase coherence?

In either case, the residual Drude peak indicates that states in the “nodal” region are not strongly impacted by the charge gap, so that the stripe-ordered state remains a nodal metal [19]. We note that the coexistence of metallic behavior and an anisotropic gap has been observed in two-dimensional CDW systems [32, 33]. Such behavior is contrary to expectations of a uniform gap for ordered stripes [11].

Before concluding, we should consider how our results compare with others in the literature. The two-component behavior (temperature-dependent Drude peak plus temperature-independent mid-infrared component) has been identified previously in studies of several different cuprate families [19, 22, 25]. The loss of spectral weight in the stripe ordered phase is a new observation. Measurements on $La_{1.275}Nd_{0.6}Sr_{0.125}CuO_4$ [12] seem to be consistent, but a larger low-temperature scattering rate in that sample masks any depression of

the optical conductivity. In contrast, studies by Lucarelli *et al.* [14] on $La_{2-x}Sr_xCuO_4$ and Ortolani *et al.* [34] on $La_{1.875}Ba_{0.125-y}Sr_yCuO_4$ have found strong, sharp peaks at finite frequency ($20-100$ cm^{-1}) in low-temperature measurements. These "far-infrared peaks", which have not been observed by other groups, have been interpreted as collective excitations of charge stripes [14, 34, 35]. Those results have been controversial, and have motivated some discussion of possible spurious effects [15, 16, 17]. Here we simply note that our measurements on a cleaved sample do not show any indications of far-infrared peaks. Furthermore, the temperature-dependence of the conductivity and the loss of the low-frequency spectral weight that we observe is directly correlated with onset of stripe order as detected by diffraction [6, 8]; the behavior of the Drude peak is quantitatively consistent with the measured resistivity.

In summary, the *ab*-plane properties of a cleaved single crystal of $La_{1.875}Ba_{0.125}CuO_4$ have been examined over a wide frequency range at several different temperatures. The rapid decrease of the carrier concentration n_D below the electronic transition at $\simeq 60$ K suggests the development of an anisotropic charge gap associated with the stripe order. Nodal excitations presumably remain ungapped; the deviation from $\rho_{ab} \sim T$ at small T is due to the decrease in carrier density, not to a uniform increase in scattering rate. Thus it appears that stripes are compatible with the nodal-metal state.

We would like to acknowledge useful discussions with D. N. Basov and T. Timusk, and helpful comments from S. A. Kivelson. This work was supported by the Office of Science, U.S. Dept. of Energy, under contract number DE-AC02-98CH10886.

* Electronic address: homes@bnl.gov
† Department of Physics, The University of Akron, Akron, OH 44325

[1] S. A. Kivelson, I. P. Bindloss, E. Fradkin, V. Oganesyan, J. M. Tranquada, A. Kapitulnik, and C. Howald, Rev. Mod. Phys. **75**, 1201 (2003).
[2] J. Zaanen, O. Y. Osman, H. V. Kruis, Z. Nussinov, and J. Tworzydło, Phil. Mag. B **81**, 1485 (2001).
[3] K. Machida, Physica C **158**, 192 (1989).
[4] M. Vojta and S. Sachdev, Phys. Rev. Lett. **83**, 3916 (1999).
[5] C. Castellani, C. Di Castro, and M. Grilli, Phys. Rev. Lett. **75**, 4650 (1995).
[6] M. Fujita, H. Goka, K. Yamada, J. M. Tranquada, and L. P. Regnault, Phys. Rev. B **70**, 104517 (2004).
[7] N. Ichikawa, S. Uchida, J. M. Tranquada, T. Niemöller, P. M. Gehring, S.-H. Lee, and J. R. Schneider, Phys. Rev. Lett. **85**, 1738 (2000).
[8] P. Abbamonte, A. Rusydi, S. Smadici, G. D. Gu, G. A. Sawatzky, and D. L. Feng, Nature Phys. **1**, 155 (2005).
[9] E. Arrigoni, E. Fradkin, and S. A. Kivelson, Phys. Rev. B **69**, 214519 (2004).
[10] S. A. Kivelson, E. Fradkin, and V. J. Emery, Nature **393**, 550 (1998).
[11] C. Castellani, C. D. Castro, M. Grilli, and A. Perali, Physica C **341-348**, 1739 (2000).
[12] M. Dumm, D. N. Basov, S. Komiya, Y. Abe, and Y. Ando, Phys. Rev. Lett. **88**, 147003 (2002).
[13] S. Tajima, N. L. Wang, N. Ichikawa, H. Eisaki, S. Uchida, H. Kitano, T. Hanaguri, and A. Maeda, Europhys. Lett. **47**, 715 (1999).
[14] A. Lucarelli, S. Lupi, M. Ortolani, P. Calvani, P. Maselli, M. Capizzi, P. Giura, H. Eisaki, N. Kikugawa, T. Fujita, et al., Phys. Rev. Lett. **90**, 037002 (2003).
[15] A. Lucarelli, S. Lupi, M. Ortolani, P. Calvani, P. Maselli, and M. Capizzi, Phys. Rev. Lett. **91**, 129702 (2003).
[16] S. Tajima, S. Uchida, D. van der Marel, and D. N. Basov, Phys. Rev. Lett. **91**, 129701 (2003).
[17] S. Tajima, Y. Fudamoto, T. Kakeshita, B. Gorshunov, V. Železný, K. M. Kojima, M. Dressel, and S. Uchida, Phys. Rev. B **71**, 094508 (2005).
[18] T. Yoshida *et al.*, Phys. Rev. Lett. **91**, 027001 (2003).
[19] Y. S. Lee, K. Segawa, Z. Q. Li, W. J. Padilla, M. Dumm, S. V. Dordevic, C. C. Homes, Y. Ando, and D. N. Basov, Phys. Rev. B **72**, 054529 (2005).
[20] C. C. Homes, M. Reedyk, D. A. Crandles, and T. Timusk, Appl. Opt. **32**, 2976 (1993).
[21] S. Uchida, T. Ido, H. Takagi, T. Arima, Y. Tokura, and S. Tajima, Phys. Rev. B **43**, 7942 (1991).
[22] F. Gao, D. B. Romero, D. B. Tanner, J. Talvacchio, and M. Forrester, Phys. Rev. B **47**, 1036 (1993).
[23] T. Startseva, T. Timusk, A. V. Puchkov, D. N. Basov, H. A. Mook, M. Okuya, T. Kimura, and K. Kishio, Phys. Rev. B **59**, 7184 (1999).
[24] W. J. Padilla, M. Dumm, S. Komiya, Y. Ando, and D. N. Basov, Phys. Rev. B **72**, 205101 (2005).
[25] D. B. Romero, C. D. Porter, D. B. Tanner, L. Forro, D. Mandrus, L. Mihaly, G. L. Carr, and G. P. Williams, Phys. Rev. Lett. **68**, 1590 (1992).
[26] A. R. Moodenbaugh, Y. Xu, M. Suenaga, T. J. Folkerts, and R. N. Shelton, Phys. Rev. B **38**, 4596 (1988).
[27] M. Ortolani, P. Calvani, and S. Lupi, Phys. Rev. Lett. **94**, 067002 (2005).
[28] The lower bound starts at zero frequency for the sum rule, and as a result the low-frequency extrapolation is included; this introduces an element of uncertainty as to the nature and position of the crossover. For instance, if the lower bound is chosen to be 10 cm^{-1}, the crossover becomes difficult to observe.
[29] A. Zimmers, J. M. Tomczak, R. P. S. M. Lobo, N. Bontemps, C. P. Hill, M. C. Barr, Y. Dagan, R. L. Greene, A. J. Millis, and C. C. Homes, Europhys. Lett. **70**, 225 (2005).
[30] X. J. Zhou, P. Bogdanov, S. A. Kellar, T. Noda, H. Eisaki, S. Uchida, Z. Hussain, and Z.-X. Shen, Science **286**, 268 (1999).
[31] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
[32] A. W. McConnell, B. P. Clayman, C. C. Homes, M. Inoue, and N. Negishi, Phys. Rev. B **58**, 13565 (1998).
[33] V. Vescoli, L. Degiorgi, H. Berger, and L. Forró, Phys. Rev. Lett. **81**, 453 (1998).
[34] M. Ortolani, P. Calvani, S. Lupi, U. Schade, A. Perla, M. Fujita, and K. Yamada (2005), cond-mat/0506018.
[35] L. Benfatto and C. Morais Smith, Phys. Rev. B **68**, 184513 (2003).